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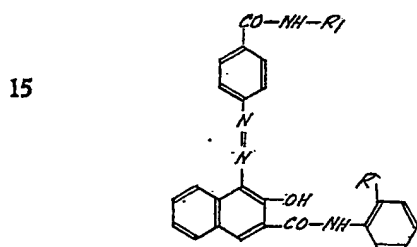
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### COMPLETE SPECIFICATION

### Water-Insoluble Monoazo Dyestuffs and processes for their manufacture and use

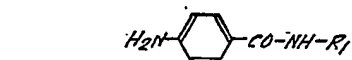
We, FARBWERKE HOECHST AKTIENGESELLSCHAFT vormals Meister Lucius & Bruning, a body corporate recognised, under German law, of 6230 Frankfurt (M)-Hoechst, Germany, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

The present invention provides new water-insoluble monoazo dyestuffs and processes for their manufacture; more particularly the invention provides dyestuffs of the following formula



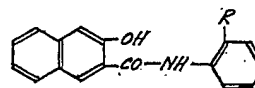
in which R represents a methyl, methoxy or ethoxy group and R<sub>1</sub> represents a hydrogen atom, an alkyl group having from 1 to 4 carbon atoms, or a phenyl or naphthyl group which may be substituted by halogen atoms, alkyl and/or alkoxy groups, and processes for their manufacture by coupling or condensation.

The invention is based on the observation that water-insoluble monoazo dyestuffs of the above general formula can be obtained when



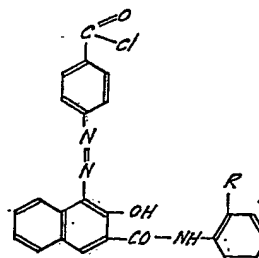
in which R<sub>1</sub> is defined as above, is diazotized and coupled with a coupling component of the general formula

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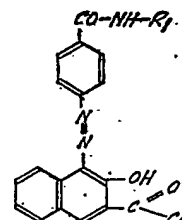


in which R is defined as above, in an aqueous medium or in the presence of an inert organic solvent, or

b) either a carboxylic acid halide of the general formula



in which R is defined as above, is condensed with ammonia or an agent yielding ammonia or with a primary aliphatic or aromatic amine, or a carboxylic acid halide of the general formula

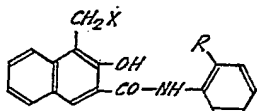


in which R<sub>1</sub> is defined as above, is condensed with 1 - amino - 2 - methylbenzene, 1 - amino - 2 - methoxybenzene or 1 - amino - 2 - ethoxybenzene.

The dyestuffs can be prepared by coupling in substance, on the fibre or on another substratum. For preparing the dyestuffs in substance 1 - aminobenzene - 4 - carboxylic acid amide, 1 - aminobenzene - 4 - carboxylic acid alkyl - amides or 1 - aminobenzene - 4 -

carboxylic acid - aryl - amides as defined above, for example 1 - aminobenzene - 4-carboxylic acid - methyl - amide, 1 - aminobenzene - 4 - carboxylic acid - ethyl - amide, 5 1 - aminobenzene - 4 - carboxylic acid - propyl - amide or 1 - aminobenzene - 4 - carboxylic acid - butyl - amide, or 1 - aminobenzene - 4 - carboxylic acid - phenyl- 10 amide or 1 - aminobenzene - 4 - carboxylic acid - naphthyl - amide which may contain as substituents halogen atoms, alkyl or alkoxy groups in the phenyl or naphthyl radical, are diazotized and coupled with 1 - (2':3'-hydroxy - naphthoylamino) - 2 - methyl- 15 benzene, 1 - (2':3' - hydroxynaphthoylamino) - 2 - methoxybenzene, or 1 - (2':3'-hydroxynaphthoylamino) - 2 - ethoxybenzene in an aqueous medium preferably in the presence of a non-ionogenic anion or cation active 20 dispersion agent or in the presence of an organic solvent, for example, pyridine, quinoline or dimethyl-formamide. When coupling in an aqueous medium, it is advantageous to heat the coupling mixture for some time so as to obtain the pigment in the optimum 25 granular condition, for example, to boil it in the presence of small quantities of an organic solvent, for example, pyridine, a hydrocarbon, a chlorohydrocarbon, e.g. dichlorobenzene, a phthalic acid dialkyl ester or a resin soap.

Mannich bases of the formula



in which R represents a methyl, methoxy or ethoxy group and X represents the radical of 35 a primary or secondary aliphatic amine which may be substituted, or the radical of piperidine or morpholine can also be used as coupling components in the process of the present invention.

40 In order to obtain particularly pure and fast dyeings it is advantageous to purify said dyestuffs prepared by coupling in substance, by stirring them for some time, if desired with heating, for example with pyridine, dimethyl- 45 formamide or another suitable organic solvent. It is advantageous in some cases to grind the crude pigment in the presence of a suitable auxiliary grinding agent, in order to convert it into a finely divided form.

50 The preparation of the dyestuffs by coupling on fibres of natural or synthetic origin, for example cotton, regenerated cellulose, polyvinyl alcohol fibre, acetate silk or poly- 55 amide fibres, is carried out according to the known ice-colour technique for dyeing and printing.

The coupling may also be carried out on other substrata for example on barium sulphate.

60 The preparation of the dyestuffs by condensation can be carried out in various ways. The carboxylic acid halide, obtained for ex-

ample, by coupling diazotized 1 - amino- 65 benzene - 4 - carboxylic acid with 1 - (2':3'-hydroxynaphthoylamino) - 2 - methylbenzene, 1 - (2':3' - hydroxynaphthoylamino)- 2 - methoxybenzene, or 1 - (2':3' - hydroxynaphthoylamino) - 2 - ethoxybenzene, and treating with an agent which converts the 70 carboxylic acid group into the carboxylic acid halide group, e.g. phosphorus trichloride, phosphorus pentachloride, phosphorous oxychloride, phosphorus pentabromide or thionyl chloride, is condensed with ammonia or agents yielding ammonia, for example ammonium 75 salts, or with primary aliphatic or aromatic amines; or the carboxylic acid halide, obtained by coupling diazotized 1 - aminobenzene- 4 - carboxylic acid amide, 1 - aminobenzene- 80 4 - carboxylic acid alkyl amides or 1 - aminobenzene - 4 - carboxylic acid aryl amides with 2:3 - hydroxy - naphthoic acid and treating with an agent which converts the 85 carboxylic acid group into the carboxylic acid halide group, is condensed with 1 - amino- 2 - methylbenzene, 1 - amino - 2 - methoxybenzene or 1 - amino - 2 - ethoxybenzene, advantageously in an inert solvent, e.g. di- 90 chlorobenzene, chlorobenzene, toluene or dioxane.

The new dyestuffs prepared by coupling 95 or condensation in substance are water-insoluble pigments which yield dyeings distinguished by a good fastness to light and to solvents. The dyestuffs are suitable for dyeing 100 or printing textile materials of vegetable or animal fibres, e.g. wool, cotton or linen or of semi-synthetic fibres, for example regenerated cellulose, artificial silk or viscose, or of 105 synthetic fibres, produced, for example, from polycondensation, polymerization or polyaddition products, according to the known pigment dyeing or pigment printing processes. The dyestuffs may also be added to spinning 110 solutions, if desired, before polycondensation or polymerization. Furthermore, they are suitable for dyeing or printing paper or paper- 115 board and for dyeing paper pulp, as well as for colouring lacquers and films of different composition, for example, of cellulose acetate, cellulose propionate or cellulose butyrate, nitrocellulose, polyvinyl acetate, polyvinyl chloride, copolymers of vinyl chloride and vinylidene chloride, polyethylene, polypropylene poly- 120 amides, polyacrylonitrile or its copolymers, polyesters or alkyd resins.

The novel dyestuffs are also suitable for 125 dyeing natural or synthetic resins, for example epoxy resins, polyester resins, vinyl resins, polystyrene resins, alkyd resins or aldehyde resins, e.g. phenol-, urea- or melamine - formaldehyde condensation products and for colouring emulsions of synthetic resins, for example, oil-in-water or water-in-oil emulsions. Furthermore they may be used for 130 dyeing natural rubber-like materials, e.g. caoutchouc or guttapercha or synthetic vulcanizable materials, e.g. polychloroprene, ole-

finic polysulphides, polybutadiene or copolymers from butadiene and styrene or materials made from butadiene and acrylonitrile.

The following Examples illustrate the invention, the parts being by weight unless otherwise stated:

#### EXAMPLE 1.

13.6 Parts of 1 - aminobenzene - 4 - carboxylic acid amide were mixed, while stirring for some time, with 60 parts by volume of 5N hydrochloric acid. The mixture was then diluted with water and diazotized at 10°C. with 20 parts by volume of 5N sodium nitrite solution. After having been clarified by treating with kieselguhr at 25—30°C., the diazonium solution thus obtained was introduced in the course of 30—45 minutes into a clarified solution of 32 parts of 1 - (2':3'-hydroxynaphthoylamino) - 2 - ethoxybenzene in 1000 parts by volume of pyridine. After the coupling, the mixture was stirred for 1 hour, the dyestuff filtered with suction and freed from the still adhering pyridine by distillation with steam, again suction-filtered, washed and dried. A bluish-red pigment dyestuff was obtained.

Graphic printings, lacquerings, polyvinyl chloride dyeings and textile dyeings made by means of this dyestuff possessed a pure bluish-red tint of good fastness to light and to solvents.

#### EXAMPLE 2.

By proceeding according to the method described in Example 1 and using 30.5 parts of 1 - (2':3' - hydroxynaphthoyl amino) - 2 - methoxybenzene or 29 parts of 1 - (2':3' - hydroxynaphthoylamino) - 2 - methylbenzene instead of 32 parts of 1 - (2':3' - hydroxynaphthoylamino) - 2 - ethoxybenzene, pigment dyestuffs were obtained, which produced dyeings with a somewhat more bluish tint, but which possessed similar fastness properties to the dyestuff described in Example 1.

#### EXAMPLE 3.

23 Parts of the monoazo - dyestuff prepared by coupling 1 mol of diazotized 1-aminobenzene - 4 - carboxylic acid with 1 mol of 1 - (2':3' - hydroxynaphthoylamino) - 2 - ethoxybenzene were heated under reflux with 300 parts by volume of chlorobenzene, and the small quantity of water present was distilled off azeotropically. The mixture was cooled to 70°C. and 2.5 parts by volume of dimethyl formamide and 5 parts by volume of thionyl chloride were added. The mixture was then gradually heated to boiling and refluxed until no more hydrochloric acid formed. The carboxylic acid chloride of the dyestuff thus obtained was filtered with suction and washed with chlorobenzene and petroleum ether. 5 Parts of the carboxylic acid chloride were heated with 250 parts by volume of dioxane and 5 parts of ammonium acetate for 2½ hours on a steam bath while stirring. The amidation of the carboxylic acid chloride set

in very rapidly. The dyestuff formed was filtered with suction, washed with water and dried. It had a similar tint to the dyestuff described in Example 1.

#### EXAMPLE 4.

13.6 Parts of 1 - aminobenzene - 4 - carboxylic acid amide were diazotized as described in Example 1. In the meantime 33 parts of 1 - (2':3' - hydroxynaphthoylamino) - 2 - ethoxybenzene were dissolved by heating with 450 parts by volume of water and 42 parts by volume of 5N sodium hydroxide solution, and the solution clarified. This solution was dropped at 10—15°C. within 30—45 minutes while stirring into the diazo solution to which 7 parts by volume of glacial acetic, 50 parts by volume of 2N sodium acetate solution and 2 parts of the reaction product of about 20 mols of ethylene oxide and 1 mol of stearyl alcohol had been added. After the coupling aqueous solutions of 5 parts of resin soap and 3 parts of calcium chloride were added and the mixture boiled for 1 hour to improve the granulation. The dyestuff thus obtained was filtered with suction, washed and dried. The dyeings produced with this dyestuff possessed practically the same tints and fastness properties as those produced with the dyestuff obtained according to Example 1.

By varying the coupling conditions, the dyestuff can be prepared in other particulate forms which show variations in tint. By coupling for example according to the following method, a dyestuff was obtained which produced distinctly more yellowish red dyeings:

33 Parts of 1 - (2':3' - hydroxynaphthoylamino) - 2 - ethoxybenzene were dissolved, as described above, in dilute aqueous sodium hydroxide solution. The clarified solution was precipitated at 0—3°C. by the addition of acetic acid in the presence of the reaction product of about 20 mols of ethylene oxide and 1 mol of stearyl alcohol and coupled at 35—40°C. and at a pH value of 6.5 with a diazo solution prepared according to the method of Example 1 from 13.6 parts of 1-aminobenzene - 4 - carboxylic acid amide. After the coupling, an aqueous solution of 5 parts of resin soap was first added to the mixture and then 3 parts of calcium chloride to improve the granular condition of the pigment. The mixture was boiled for 1 hour and the dyestuff filtered with suction, washed and dried.

#### EXAMPLE 5.

14 Grams of 1 - (2':3' - hydroxynaphthoylamino) - 2 - methoxybenzene were dissolved by boiling in 1 litre of soft water which contained 14 cc of 32.5% sodium hydroxide solution and 20 cc of an oil sulphonate wetting agent. A cotton fabric was immersed into the afore-described solution and squeezed between rollers to obtain a liquid absorption of 100% by weight of the goods to be treated.

It was then dried by means of hot air or contact heat.

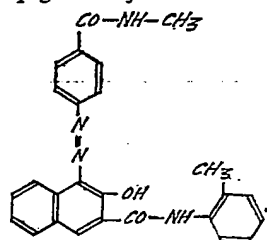
The fabric impregnated in this way was then immersed into the following bath and again squeezed between rollers to obtain a liquid absorption of 100% by weight of the goods to be treated:

7.5 Grams of 1 - aminobenzene - 4 - carboxylic acid amide were diazotized as described in Example 1 and stirred into 1 litre of water which contained 1 gram of the reaction product of about 20 mols of ethylene oxide and 1 mol of octadecyl alcohol and which, together with the acid of the diazotization bath, contained enough acetic acid to neutralize the 14 cc of 32.5% sodium hydroxide solution used as above (18.5 cc of 50% acetic acid). The fabric was then rinsed in water, treated for 15 minutes at 95°C. with 1 gram of the reaction product of about 10 mols of ethylene oxide and 1 mol of nonyl phenol and 3 grams of sodium carbonate per litre of water, rinsed and dried.

A red dyeing possessing good fastness properties was obtained.

#### EXAMPLE 6.

15 Parts of 1 - aminobenzene - 4 - carboxylic acid - methyl - amide were stirred for 15 minutes with 150 parts by volume of water and 75 parts by volume of 5N hydrochloric acid and then diazotized with 20 parts by volume of 5N sodium nitrite solution. The clarified diazo solution was introduced into a solution of 29 parts of 1 - (2':3' - hydroxynaphthoylamino) - 2 - methylbenzene in 750 parts by volume of pyridine in the course of about 30 minutes. After the coupling, stirring was continued for about a further 30 minutes and the dyestuff filtered with suction, washed thoroughly with water and dried at 60°C. The pigment dyestuff of the formula



was obtained which produced intense orange-red dyeings and possessed good fastness properties when incorporated into printing inks or spinning masses.

#### EXAMPLE 7.

The coupling was carried out according to the method described in Example 6, but instead of 29 parts of 1 - (2':3' - hydroxynaphthoylamino) - 2 - methylbenzene 30.5 parts of 1 - (2':3' - hydroxynaphthoylamino) - 2 - methoxybenzene were used. A pigment

dyestuff was obtained which produced scarlet-red dyeings. Its fastness properties corresponded to those of the dyestuff described in Example 6.

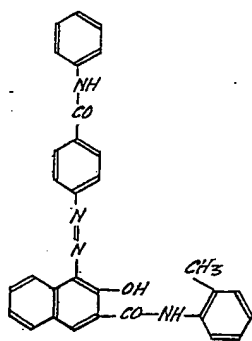
#### EXAMPLE 8.

21.5 Parts of the monoazo - dyestuff prepared by coupling 1 mol of diazotized 1-aminobenzene - 4 - carboxylic acid with 1 mol of 1 - (2':3' - hydroxynaphthoylamino) - 2 - methyl - benzene were heated under reflux with 300 parts by volume of chlorobenzene, and the small quantity of water present was distilled off azeotropically. The mixture was cooled to 70°C. and 2.5 parts by volume of dimethyl - formamide and 5 parts by volume of thionyl chloride were added. The mixture was then gradually heated to boiling and refluxed until no more hydrochloric acid formed. The carboxylic acid chloride of the dyestuff thus obtained was filtered with suction and washed with chlorobenzene and petroleum ether. 4.5 Parts of the carboxylic acid chloride were heated under reflux for about 5 hours with 200 parts by volume of chlorobenzene, 2 parts by volume of pyridine and 1 part of aniline. The dyestuff formed was then filtered with suction, washed with petroleum ether and hot water and dried. It was an orange-red powder.

#### EXAMPLE 9.

31.8 Parts of 1 - aminobenzene - 4 - carboxylic acid - phenyl - amide were introduced into a mixture of 150 parts by volume of water and 75 parts by volume of 5N hydrochloric acid and stirred for some hours. The mixture was then diluted with 1500 parts by volume of water and diazotized with 30 parts by volume of 5N sodium nitrite solution.

43 parts of 1 - (2':3' - hydroxynaphthoylamino) - 2 - methylbenzene were introduced into 350 parts by volume of water and 36 parts by volume of 5N sodium hydroxide solution and dissolved by heating to 75°C. The hot solution was cooled to room temperature by the addition of ice and introduced into a mixture of 500 parts by volume of water, 20 parts by volume of glacial acetic acid and 3 parts of a wetting agent mixture consisting of an alkyl sulphonate and a hydroxyethylated fatty alcohol. A white suspension was obtained which was heated to 60—65°C. and at this temperature the clarified diazo solution was introduced into the suspension, the coupling mixture being maintained acidic with acetic acid by the addition of a sodium acetate solution. After the coupling, 3 grams of resin soap were added and the mixture boiled for a further hour. After filtering with suction, washing and drying, an orange-red dyestuff of the formula



was obtained.

This dyestuff when incorporated into a graphic printing ink gave brilliant orange-red dyeings of an intense colour and good fastness properties.

#### EXAMPLE 10.

14 Grams of 1 - (2':3' - hydroxynaphthoyl-amino) - 2 - methyl - benzene were dissolved by boiling in 1 litre of soft water which contained 14 cc of 32.5% sodium hydroxide solution and 20 cc of an oil sulphonate wetting agent. A cotton fabric was immersed into the afore-described solution and squeezed between rollers to obtain a liquid absorption of 100% by weight of the goods to be treated. The fabric was then dried by means of hot air or contact heat. The fabric thus impregnated was then immersed in the following

bath and again squeezed between rollers to obtain a liquid absorption of 100% by weight of the goods to be treated:

9 Grams of 1 - aminobenzene - 4 - carboxylic acid ethyl amide were diazotized as described in Example 6 and stirred into 1 litre of water which contained 1 gram of the reaction product of about 20 mols of ethylene oxide and 1 mol of octadecyl alcohol and which together with the acid of the diazotization bath contained enough acetic acid to neutralize the 14 cc of 32.5% sodium hydroxide solution referred to above. The fabric was then rinsed in water, treated for 15 minutes at 95°C. with 1 gram of the reaction product of about 10 mols of ethylene oxide and 1 mol of nonyl phenol and 3 grams of sodium carbonate per litre of water. Subsequently it was rinsed and dried. A scarlet-red dyeing possessing good fastness properties was obtained.

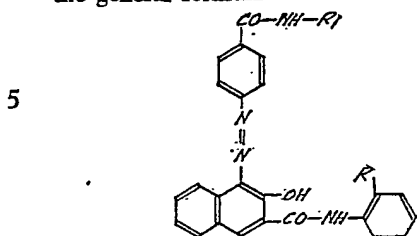
If instead of 9 grams of 1 - aminobenzene-4 - carboxylic acid ethyl amide the equivalent quantity of 1 - aminobenzene - 4 - carboxylic acid butyl amide was used, a somewhat more yellowish dyeing was obtained which possessed the same good fastness properties.

The following table contains further components which can be used according to the present invention and the tints produced with the dyestuffs obtained:

Diazo component	Coupling component	Tint
1-aminobenzene-4-carboxylic acid-(2'-chloro)-phenyl-amide	1-(2':3'-hydroxy-naphthoylamino)-2-ethoxybenzene	red
1-aminobenzene-4-carboxylic acid-(2':4'-dichloro)-phenyl-amide	"	"
1-aminobenzene-4-carboxylic acid-(2'-methyl-3'-chloro)-phenyl-amide	1-(2':3'-hydroxy-naphthoylamino)-2-methylbenzene	orange-red
1-aminobenzene-4-carboxylic acid-(2'-methoxy)-phenyl-amide	"	"
1-aminobenzene-4-carboxylic acid-(2'-methyl)-phenyl-amide	"	"
1-aminobenzene-4-carboxylic acid-(2'-methyl-5'-chloro)-phenyl-amide	"	"
1-aminobenzene-4-carboxylic acid-(2':5'-dichloro)-phenyl-amide	"	"
1-aminobenzene-4-carboxylic acid-(3':4'-dichloro)-phenyl-amide	"	"
1-aminobenzene-4-carboxylic acid-(2'-methoxy-5'-chloro)-phenyl-amide	"	"
1-aminobenzene-4-carboxylic acid-(2':4'-dimethyl)-phenyl-amide	1-(2':3'-hydroxy-naphthoylamino)-2-methoxybenzene	"
1-aminobenzene-4-carboxylic acid- $\alpha$ -naphthyl-amide	1-(2':3'-hydroxy-naphthoylamino)-2-methylbenzene	"
1-aminobenzene-4-carboxylic acid- $\beta$ -naphthyl-amide	"	"

## WHAT WE CLAIM IS:—

1. Water-insoluble monoazo dyestuffs of the general formula



in which R represents a methyl, methoxy or ethoxy group and R<sub>1</sub> represents a hydrogen atom or an alkyl group having from 1—4

carbon atoms or a phenyl or naphthyl group which may be substituted by halogen atoms, alkyl and/or alkoxy groups. 10

2. Any one of the water-insoluble monoazo dyestuffs obtainable as described in the Examples or with reference to the Table herein. 15

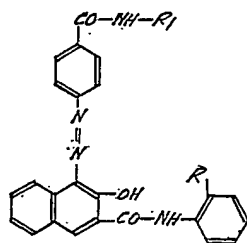
3. A dyeing, colouring, or printing process wherein a water-insoluble monoazo dyestuff claimed in Claim 1 or 2 is used. 15

4. A process for dyeing, printing, or colouring as claimed in Claim 3 conducted substantially as described and exemplified herein. 20

5. Vegetable, animal, or synthetic textile fibrous materials, spinning solutions, paper,

lacquers and films which have been dyed, printed, or coloured by a process claimed in Claim 3 or 4.

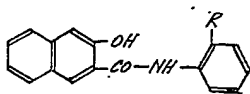
- 5 6. A process for the manufacture of water-insoluble monoazo dyestuffs of the general formula



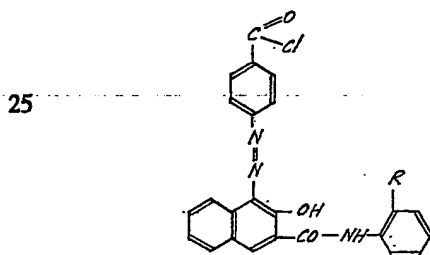
- 10 in which R represents a methyl, methoxy or ethoxy group and R<sub>1</sub> represents a hydrogen atom or an alkyl group having from 1 to 4 carbon atoms, or a phenyl or naphthyl group which may be substituted by halogen atoms, alkyl and/or alkoxy groups wherein  
a) an amine of the general formula



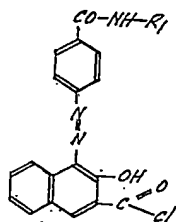
in which R<sub>1</sub> is defined as above, is diazotized and coupled with a coupling component of the general formula



- 20 in which R is defined as above, in an aqueous medium or in the presence of an inert organic solvent, or  
b) either a carboxylic acid halide of the general formula



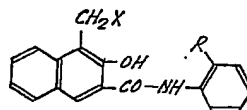
- 30 in which R is defined as above, is condensed with ammonia or an agent yielding ammonia or with a primary aliphatic or aromatic amine, or a carboxylic acid halide of the general formula



in which R<sub>1</sub> is defined as above, is condensed with 1 - amino - 2 - methylbenzene, 1 - amino - 2 - methoxybenzene or 1 - amino - 2 - ethoxybenzene.

7. A process as claimed in Claim 6, wherein an amine of the general formula given, is diazotized and coupled on the fibre or on a substratum and with a coupling component of the general formula given.

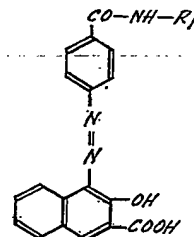
8. A process for the manufacture of water-insoluble monoazo dyestuffs according to Claim 1 wherein a 1 - aminobenzene - 4 - carboxylic acid of the general formula given is diazotized and coupled in substance, on the fibre, or on a substratum with a coupling component of the general formula



in which R represents a methyl, methoxy or ethoxy group and X represents the radical of a primary or secondary aliphatic amine which may be substituted, or the residue of piperidine or morpholine.

9. A process as claimed in Claim 6, wherein diazotized 1 - aminobenzene - 4 - carboxylic acid is coupled with 1 - (2':3'-hydroxynaphthoylamino) - 2 - methylbenzene, -2 - methoxybenzene, or -2 - ethoxybenzene, the carboxylic acid group of the azo dyestuff so obtained is transformed into the corresponding carboxylic acid chloride group by known methods and the compound so obtained is condensed with ammonia, a compound yielding ammonia or with a primary aliphatic or aromatic amine.

10. A process as claimed in Claim 6, wherein a monoazo dyestuff of the general formula



in which R<sub>1</sub> has the meaning given in Claim 6, is transformed into the corresponding carboxylic acid chloride by known methods and the acid chloride is condensed with 1 - amino - 2 - methylbenzene, 1 - amino - 2 - methoxybenzene or 1 - amino - 2 - ethoxybenzene.

11. A process for the manufacture of water-insoluble monoazo dyestuffs as claimed in Claim 1 conducted substantially as described in any one of the Examples or with reference to the Table herein.

12. A process for the manufacture of water-insoluble monoazo dyestuffs as

claimed in Claim 1 substantially as described herein.

13. Water-insoluble monoazo dyestuffs when  
obtained by a process claimed in any one of  
5 Claims 6—12.

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